

Two-dimensional frustrated spin systems in high magnetic fields

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Abstract. We discuss our numerical results on the properties of the $S = 1/2$ frustrated J_1 - J_2 Heisenberg model on a square lattice as a function of temperature and frustration angle $\phi = \tan^{-1}(J_2/J_1)$ in an applied magnetic field. We cover the full phase diagram of the model in the range $-\pi \leq \phi \leq \pi$. The discussion includes the parameter dependence of the saturation field itself, and addresses the instabilities associated with it. We also discuss the magnetocaloric effect of the model and show how it can be used to uniquely determine the effective interaction constants of the compounds which were investigated experimentally.

1. Introduction

The ground-state and low-temperature properties of low-dimensional frustrated spin systems are of great interest due to their unusual and sometimes unexpected temperature and magnetic-field dependence. The origin for this lies in the enhanced quantum fluctuations which determine their behaviour. A prime example for such a system is the spin-1/2 frustrated J_1 - J_2 Heisenberg model on a square lattice, being one of the most intensively studied models for spin systems. It has a Hamiltonian of the form

$$H = J_1 \sum_{\langle ij \rangle_1} \mathbf{S}_i \mathbf{S}_j + J_2 \sum_{\langle ij \rangle_2} \mathbf{S}_i \mathbf{S}_j, \quad (1)$$

where J_1 denotes the exchange interaction between neighbouring spins, and J_2 labels the exchange interaction along the diagonals between next nearest neighbours.

In order to facilitate the description of the model and its thermodynamics, it is convenient to introduce two parameters to characterise it, namely an overall energy scale J_c and a frustration angle ϕ defined by

$$J_c = \sqrt{J_1^2 + J_2^2} \quad \text{and} \quad \phi = \tan^{-1}(J_2/J_1), \quad (2)$$

such that $J_1 = J_c \cos \phi$, and $J_2 = J_c \sin \phi$. Except where stated otherwise, the numerical results discussed in the following are all obtained using the finite-temperature Lanczos method [1] on a 20-site square with periodic boundary conditions.

2. Phase diagram and experiments

Figure 1 shows the phase diagram of the model, together with some extra lines referring to the compounds which were investigated experimentally. At a classical level, the phase diagram has

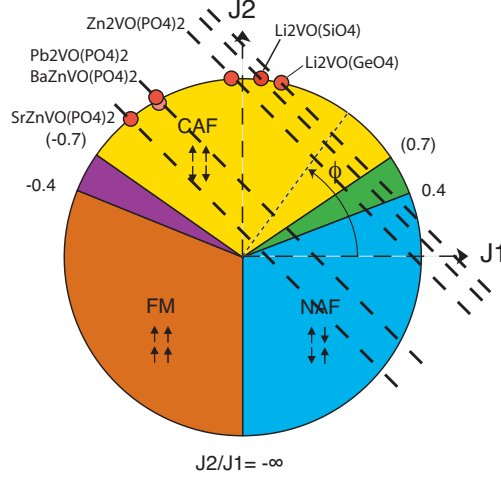


Figure 1. Schematic phase diagram of the J_1 - J_2 model. The boundary between FM and NAF phase is the line $J_1 = 0$, $J_2 < 0$. Numbers are ratios of exchange couplings J_2/J_1 where zero point fluctuations destroy the relevant order parameter [1, 2]. The dashed lines labeled by the chemical formulae for the experimentally known “ J_1 - J_2 compounds” are lines of constant $\Theta_{\text{CW}} = (J_1 + J_2)/k_B$ as determined from $H = 0$ heat capacity and magnetic susceptibility measurements [3, 4].

three main phases: For $-\pi/2 < \phi < \tan^{-1}(1/2)$, the Néel antiferromagnet (NAF) characterised by an ordering vector $\mathbf{Q} = (\pi, \pi)$, a ferromagnetic phase (FM) for $\pi - \tan^{-1}(1/2) < \phi < -\pi/2$, and a collinear phase (CAF) with $\mathbf{Q} = (\pi, 0)$ or $(0, \pi)$ for $\tan^{-1}(1/2) < \phi < \pi - \tan^{-1}(1/2)$. At the boundary between CAF and NAF, the Ising domain wall energy between the states vanishes, and arbitrary stripes of the two phases can be formed. At the CAF/FM boundary, coplanar spiral states with $\mathbf{q} = (2\pi n/m, 0)$, where $\{n, m\}$ are integers, all become degenerate. Quantum fluctuations lift these degeneracies and lead to disordered regions of finite width at these two boundaries having continuous classical degeneracies. A more detailed discussion of the properties of the model at zero magnetic field is given in References [1, 2] and the references cited therein.

The J_1 - J_2 model is appropriate for the description of a number of quasi two-dimensional vanadium compounds. They can be grouped in two families of type Li_2VOXO_4 ($X = \text{Si}, \text{Ge}$), and $AA'\text{VO}(\text{PO}_4)_2$ ($A, A' = \text{Pb}, \text{Zn}, \text{Sr}, \text{Ba}$), respectively [3, 4]. All compounds feature V^{4+} ions with $S = 1/2$ surrounded by oxygen polyhedra, forming layers of J_1 - J_2 square lattices which are weakly coupled in the third spatial dimension.

Thermodynamic coefficients have been measured to determine the effective interaction constants [3, 4]. For zero magnetic field, this is not completely possible. An ambiguity remains when fitting the experimental $\chi(T)$ and $C_V(T)$ using a high-temperature series expansion: We always have *two* possible points $\phi = \phi_{\pm}$ in the phase diagram symmetric around $\phi = \pi/4$. This situation does *not* change qualitatively when including higher-order terms in the expansion used for the fits [5].

3. Finite fields

Fortunately, all J_1 - J_2 compounds known so far have values $J_c = \mathcal{O}(10\text{K})$, which corresponds to a magnetic-field range accessible in pulsed high-magnetic field experiments.

We have calculated the saturation field of the magnetisation as a function of the frustration angle ϕ and the magnetic field H . The left-hand side of Figure 2 shows the result for a 20-site

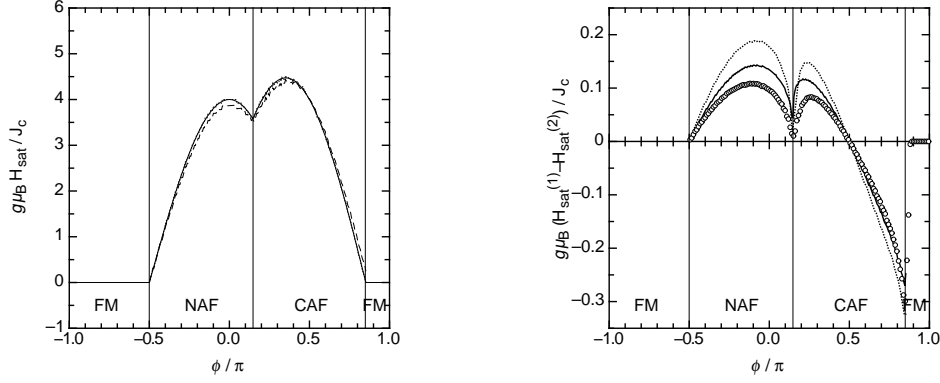


Figure 2. Left: One-magnon instability (solid line) and two-magnon instability (dashed line) field H of the fully polarized state of the J_1 - J_2 model on a 20-site square as a function of the frustration angle $\phi = \tan^{-1}(J_2/J_1)$. The one-magnon instability field corresponds to the saturation field of the magnetisation obtained by linear spinwave theory. The energy unit is $J_c = \sqrt{J_1^2 + J_2^2}$. Right: Difference between the $\Delta S = 1$ and $\Delta S = 2$ instabilities as a function of the frustration angle for a cluster with 16 sites (dotted line), 20 sites (solid line), and 24 sites (open circles).

cluster with periodic boundary conditions as well as the saturation field obtained from linear spinwave theory. We define the k^{th} saturation field as the instability of the fully polarised (saturated) state against an excitation with k magnons,

$$g\mu_B H_{\text{sat}}^{(k)} = \frac{1}{k} \left[E \left(S_{\text{tot}}^z = \frac{N}{2} \right) - E \left(S_{\text{tot}}^z = \frac{N}{2} - k \right) \right], \quad (3)$$

where E denotes the ground-state energy per site of the cluster of size N , and S_{tot}^z is the component of the total spin of the system in the direction of the magnetic field H . In this notation, the thermodynamic saturation field of the magnetisation is given by $H_{\text{sat}} = \max_k (H_{\text{sat}}^{(k)})$. The result from linear spinwave theory for the infinitely large system reads

$$g\mu_B H_{\text{sat}}^{(\text{cl})} = zS J_c \left[\cos \phi \left(1 - \frac{1}{2} (\cos Q_x + \cos Q_y) \right) + \sin \phi \left(1 - \cos Q_x \cos Q_y \right) \right] \quad (4)$$

with $z = 4$ and $S = 1/2$, and is displayed as solid line in Figure 2 (left). It is in perfect agreement with the numerical result for $H_{\text{sat}}^{(1)}$.

For values $-\pi/2 < \phi < \pi/2$ ($J_1 > 0$), the saturation field is determined by the $\Delta S = 1$ instability. Inside the collinear phase, at the point $\phi = \pi/2$, J_1 vanishes, and the lattice decouples into two sublattices. Therefore, the $\Delta S = 1$ and $\Delta S = 2$ spin flip energies must have the same value. For ferromagnetic J_1 , for the finite-size clusters considered, the $\Delta S = 2$ instability determines the saturation field until the point $\phi \approx 0.9\pi$ where the ferromagnetic phase is reached. This is seen from Figure 2 (right), where we have plotted the difference $H_{\text{sat}}^{(1)} - H_{\text{sat}}^{(2)}$ for three different cluster sizes. While the overall size of $H_{\text{sat}}^{(1)} - H_{\text{sat}}^{(2)}$ decreases monotonically with the size of the cluster, at least in the NAF and CAF phases, the crossover described above happens always exactly at $J_1 = 0$ independent of the cluster size.

4. Magnetocaloric effect

The magnetocaloric effect (MCE) is the adiabatic temperature change of a sample by applying a magnetic field, given by

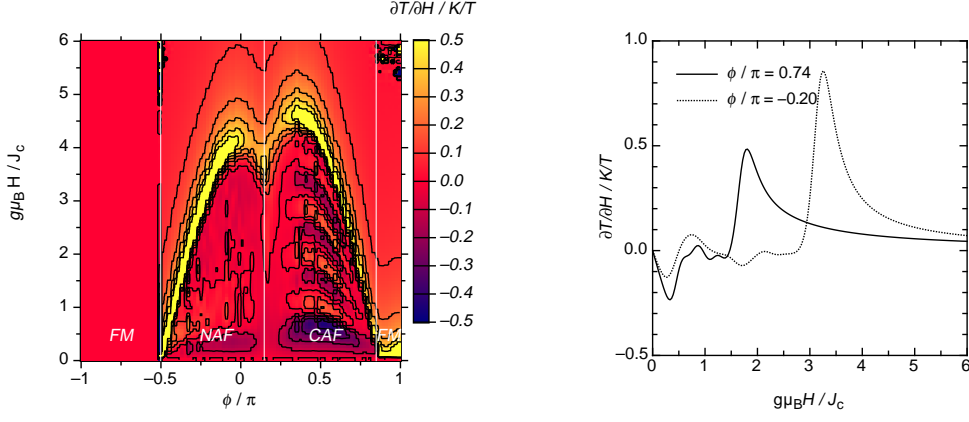


Figure 3. Left: H - ϕ contour plot of the MCE of the J_1 - J_2 model at $T = 0.2J_c/k_B$ on a 20-site square with periodic boundary conditions. The calculation was done on a grid of 330×200 points. Right: Field dependence of the MCE for those two particular values of $\phi = \phi_{\pm}$ determined for the compound $\text{SrZnVO}(\text{PO}_4)_2$ [3], again at a temperature $T = 0.2J_c/k_B$. Note the large difference of the maximum positions.

$$\left(\frac{\partial T}{\partial H}\right)_S = -\left(\frac{\partial S}{\partial H}\right)_T / \left(\frac{\partial S}{\partial T}\right)_H = -\frac{T(H)}{C_V(H, T)} \left(\frac{\partial M(H, T)}{\partial T}\right)_H, \quad (5)$$

where S is the entropy, and M the magnetisation. We have calculated the MCE on a 20-site square as a function of T , H , and ϕ . Figure 3 (left) shows the MCE for fixed temperature $T = 0.2J_c/k_B$. Its main feature is the pronounced anomaly near the saturation field where the fully polarised state is reached, corresponding to the sudden entropy change taking place upon reaching saturation. The saturation magnetisation is reached slightly above the maximum position of the MCE. The oscillating structure of the MCE below H_{sat} in the CAF phase is a finite-size effect which we expect to vanish in the thermodynamic limit. Note that in the Néel as well as in the collinear phase, the MCE is for some range of fields *negative*, indicating that the sample will *cool down* with increasing field, opposite to what is expected for simple paramagnets.

The enhanced quantum fluctuations due to the frustration are evidenced by an enhanced magnetocaloric effect in the disordered region between the CAF and the FM phases, and close to the region between the CAF and the NAF phases.

The right-hand side of Figure 3 shows two vertical cuts through the left-hand side plot for those values $\phi = \phi_{\pm}$ which have been determined experimentally from zero-field heat capacity and susceptibility measurements of the compound $\text{SrZnVO}(\text{PO}_4)_2$ [3]. In contrast to the zero-field data, the two maximum positions of the MCE anomalies differ by roughly a factor two.

The maximum saturation field of the J_1 - J_2 model is reached at $\phi_{\text{max}}/\pi = \tan^{-1}(2)/\pi \approx 0.35$ with $g\mu_B H_{\text{sat}}^{\text{max}} = 4.5J_c$. Since the available compounds all have values $J_c = \mathcal{O}(10 \text{ K})$, their saturation fields have values $H_{\text{sat}} < 40 \text{ T}$, assuming $g = 2$. These fields are accessible; therefore, a measurement of the field dependence of the magnetocaloric effect can help to clarify the nature of the ground state of the J_1 - J_2 compounds discussed here.

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